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Study of Ammonia Source at a Portland Cement Production Plant (Journal Version)

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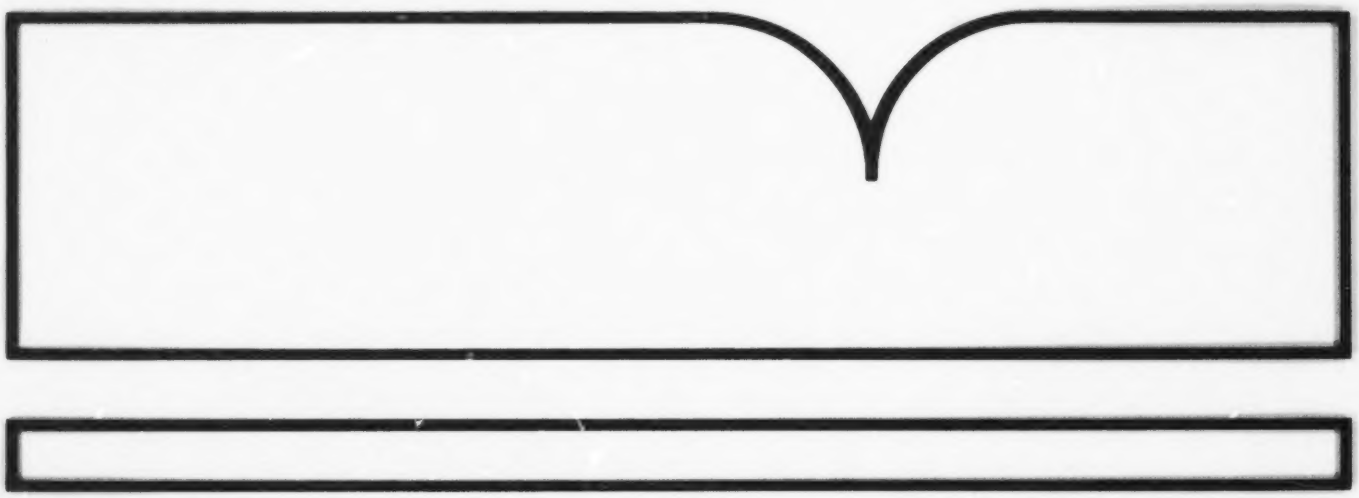

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16. ABSTRACT

A source and process sampling study was conducted at a dry process Portland Cement production plant. One aspect of the study focused on the source or point of NH_3 within the production process. An extensive number of process solids from raw feeds to baghouse solids were collected and analyzed for NH_4^+. Samples were analyzed for NH_4^+ both by washing the solids with 0.1N H_2SO_4 and by collecting NH_3 in impingers as it was evolved from heated solids.

The results show the presence of NH_4^+ in many process samples, and that the collection efficiency of NH_4^+ in the baghouse is related to baghouse temperature. The data also show that NH_3 is derived from the shale used in the raw feed at this cement production plant.

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JOURNAL ARTICLE

A Study of Ammonia Source at a Portland Cement Production Plant

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A source and process sampling study was conducted at a dry process Portland Cement production plant. The study was performed to determine the nature of the formation of a highly visable plume related to the kiln emissions. One aspect of the study focused on the source or point of NH_3 within the production process. An extensive number of process solids from raw feeds to baghouse solids were collected and analyzed for NH_4^+. Samples were analyzed for NH_4^+ both by washing the solids with 0.1 N H_2SO_4 and by collecting NH_3 in impingers as it was evolved from heated solids. The results showed that NH_4^+ was present in many process samples and that the collection efficiency of NH_4^+ in the baghouse was related to baghouse temperature. The data also showed that NH_3 was derived from the shale used in the raw feed at this cement production plant.

The presence of gaseous ammonia (NH_3) in the emissions of some Portland Cement production plants continues to result in both visible plume formation[1,2] and particulate sampling method difficulties.[3] Chadbourne *et al.*[3] have presented data which showed the effects that NH_3 can have on sampling the kiln emissions for particulate matter. That work showed how increasing the probe temperature during sampling resulted in an increase of NH_3 in the impinger catches. However, Chadbourne did not present data on total NH_3 emissions for impingers, probe and filter; and the source of NH_3 was not discussed.

Work by Griffin *et al.*[4] looked more extensively at the presence and collection of NH_3 in particulate samples collected at a cement production plant. Griffin, as did Chadbourne, confirmed the presence of NH_3 in the emissions by the NH_3 collected in the impingers. Griffin concluded that ammonium particulate and stack temperature were considered to be more a measure of the roller mill operation. In this work, as in Chadbourne's work, the presence of NH_4^+ in the particulate fraction collected in the probe and filter and the source of NH_3 were not reported.

Gartner and Wilks[5] also discussed the presence of nitrogenous compounds in cement kiln emissions. Their work addressed the potential sources of the nitrogenous compounds that could form NO_x by analyzing feedstocks with a Kjeldahl analysis. The work involved measuring NO_x that was emitted from various feedstocks when heated to 1000°C, and like previous studies did not isolate and identify the source of NH_3 from the feedstocks.

These works leave little doubt that the presence of NH_3 in cement kiln emissions can be significant and that it has an effect on particulate sampling results. However, the results of these works leave the question of the source or cause of NH_3 in the emissions unanswered. Identifying the origin of the NH_3 in the Portland Cement manufacturing process could be a significant step towards a control approach that would prevent particle formation in the plume.

One possible source of the NH_3 is that it is produced from the nitrogen in the kiln where high temperatures and adequate catalyst exist for fuel nitrogen reduction. Another possible source is that since most facilities use the conventional ammonium nitrate type blasting powder to mine the limestone, the NH_4^+ could be coming from the "residual" powder in limestone. Evolution of NH_3 from urea, protein materials and other organic nitrogen products in the raw feeds is also possible.

Recently, we conducted a source and process sampling program at a Portland Cement kiln to determine the cause of a detached high opacity plume.[6] During the study, measurements and analysis were performed to verify or dismiss some of the NH_3 ideas.

The plant sampled was the South Dakota Cement Company located at Rapid City, South Dakota. The process studied was the plant's dry process, a coal-fired kiln which produces a nominal 420,000 tons/yr of product. As is typical of most dry processes, the plant can operate either in the mill mode, when the kiln emissions are passed through the raw feed pulverizing mill prior to the baghouse, or in the bypass mode when the raw feed mill is inoperative and the kiln emissions bypass the mill and go directly to the baghouse. The opacity of the emissions instack is usually 10 percent or less while the actual particulate loading is questionable since the values obtained depend on the sampling method. Plant operators felt that improvements in the baghouse, now used to control the particulate matter, provided the capability for maintaining compliance status. The questions raised on the variability of the actual amounts of the particulate emissions arose when the study results showed significant and varying amounts of NH_4^+ and Cl^- ions in the particulate samples. For two of the samples collected during the study, nearly 55 and 67 percent of the total particulate catch was found to be NH_4Cl by chemical analysis.

Since NH_4Cl was present at significant levels in the emissions, a laboratory study was conducted to establish the source of the NH_3 at this cement plant. Three potential sources were considered: 1) residual NH_4^+ in limestone, 2) the formation of NH_3 in the kiln from nitrogen reducing chemistries, and 3) the formation or evolution of NH_3 from the other feeds. This report discusses the laboratory study of the process samples and the conclusions drawn from the study results.

Experimental

Source and process sampling was conducted at the South Dakota facility for two weeks in May 1985. Extensive gas and solids sampling was conducted to determine whether NH_3 and NH_4^+ were present at various points in the

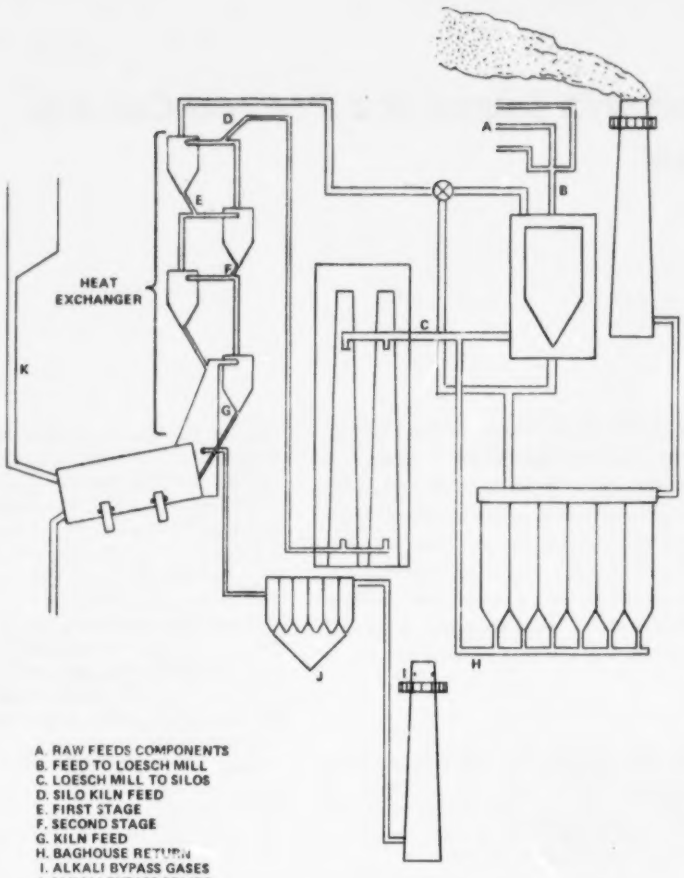

Figure 1. Schematic of the cement plant sampling points.

process. A schematic depicting the process with designated sampling points is shown in Figure 1.

To check the extent to which the blasting powder residue may contribute NH_4^+ to the process, grab samples were acquired at the limestone blasting area. Four samples were taken: two near the remaining ledge after blasting, and two more remote from this area as background samples. The samples, fragments no longer than ½ in., were placed in preweighed polyethylene containers. The containers were reweighed at the plant and then the solids were covered with 0.1 N H_2SO_4. The samples were then stored on ice for return to the laboratory for analysis.

Solid samples of the raw shale were also taken at point A (Figure 1) and of the combined raw feed going to the Loesch Mill at point B. These solid samples were also placed in polyethylene containers and stored on ice for the return to the laboratory. No acid solution was placed in these containers with the solids.

The same procedure was followed (without acid) for sampling point H, the baghouse solids returned to the silos, points D, E, F, and G the kiln feed at various points through the heat exchanger, the coal sample taken at point K, the alkali bypass solids sample at point J, and the ground raw product from the Loesch Mill to the silos.

Several gas samples were taken from the alkali bypass stack, (point I), and at the heat exchanger points E and F. The samples were collected by pulling the gas through a quartz probe plug and then through a 0.1 H_2SO_4 impinger solution. The transfer line washes were combined with the impinger catches and analyzed for NH_4^+.

Solution analysis for NH_4^+ was performed with an automated Technicon Auto-Analyzer II Colorimetric procedure.[7] Samples were taken during both operating modes for all sampling points except A, B, K, I, and J.

Routine temperatures of the operation were recorded as well as the temperature of the solid samples at acquisition times.

Two types of NH_4^+ recovery procedures were used on the solid samples. The first procedure involved washing a weighed solid sample with three 25-mL aliquots of 0.1 N H_2SO_4, and after filtering the washings, diluting to 100 mL for analysis. The second procedure involved evolving NH_3 from solid samples by heating the solid samples. For this procedure a special apparatus was used. The apparatus as shown in Figure 2 involved an airtight stainless steel container that could be heated in a muffle furnace to temperatures of 316°C to 815°C, comparable to those of the plant process. By using the procedure any NH_3 liberated by the heating was collected in a conventional impinger system containing 0.1 N H_2SO_4. Aliquots from the impinger catches were then analyzed for NH_4^+. This scheme removed any atmospheric NH_3 and collected NH_3 with no interference from nitrites or nitrates.

Results

The results of the limestone mine samples and raw shale and combined raw feed samples are shown in Table I. Very little NH_4^+ was found in the limestone collected in the limestone quarry. A slightly higher amount of NH_4^+ (0.60 ppm) was seen in the blasting samples than in the background samples (0.16 ppm) but neither was significant. However, a high amount of NH_3 was found in the shale. The four samples collected have an average of a 6.18 ± 1.2 ppm of NH_4^+ over 10 times that in the limestone. Assuming 80 and 20 percent feeds of limestone and shale respectively, this indicates that in excess of 80 percent of the acid extractable NH_3 available in the raw feed components is

Table I. Analysis of feed samples for NH_4^+.

Sample ID	Sample wt (g)	μg NH_4^+/g (ppm)
Mine (blasting zone)	131	0.54
Mine (blasting zone)	134	0.66
Mine (background)	144	0.16
Mine (background)	124	0.17
Shale	10.4	7.31
Shale	17.8	4.67
Shale	14.2	5.28
Shale	12.9	7.44
Raw feed	24.2	2.31
Raw feed	25.3	1.94
Raw feed	24.4	2.01
Raw feed	27.4	1.82

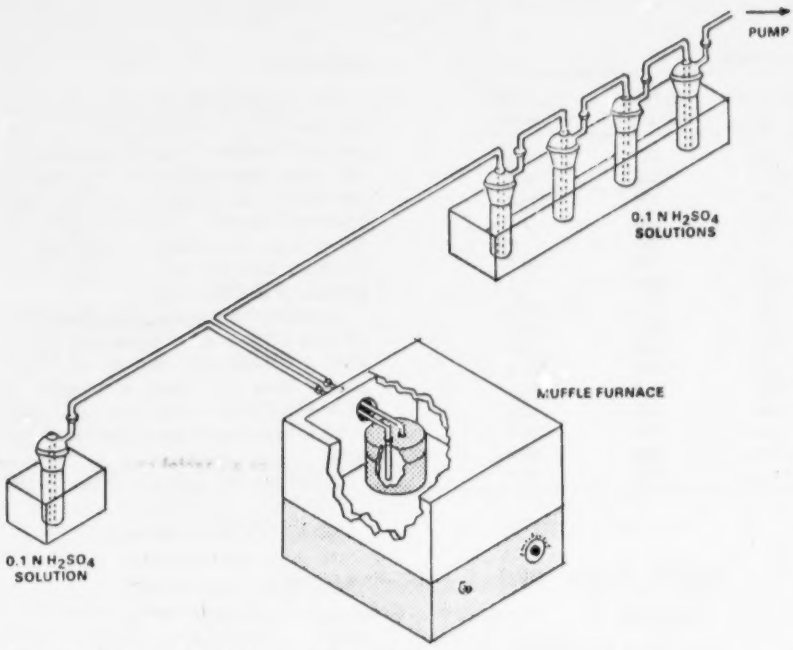

Figure. 2. Apparatus for evolution and collection of NH_3.

from the shale. Further discussion of the shale is provided later.

Analysis of the Loesch Mill solids by the acid washing procedure gave the results shown in Table II. There is no apparent NH_4^+ concentration pattern with time; however, the difference in high and low values are large for the 15 samples ranging from 23.9 to 49.1 with an average of 33.4 ppm. The presence of higher amounts of NH_4^+ in the mill sample relative to raw products is likely due to absorption of gaseous NH_3 in the mill solids. Similar sample collections and analyses were performed on the kiln feed into the heat exchanger. The results of these samples are shown in Table III.

Similar to the Loesch Mill, the kiln feed shows no relationship between NH_4^+ concentration and time. The average kiln feed NH_4^+ concentration of the solids for the mill on mode is 43.0 ± 6.5 ppm. For the mill off mode the average NH_4^+ content is 39.4 ± 7.3 ppm. No statistical difference between the two modes for the presence of NH_4^+ in the kiln feed was found. The additional NH_4^+ present in the kiln feed compared to that of the Loesch Mill is probably caused by return of the baghouse solids to the storage silos.

The results of NH_4^+ analysis for the baghouse solids returned to the silos is shown in Table IV. An average 129 ± 20 ppm of NH_4^+ was present in the baghouse solids collected during the mill on mode, and an average 73.9 ppm NH_4^+ was present in the bypass mode. The apparent difference can be attributed to either the collection of NH_3 in the Loesch Mill and subsequent collection in the baghouse or to the lower temperature of the baghouse during this operational mode which would enhance particulate NH_4Cl formation. That less NH_4^+ was collected in the baghouse during the higher temperatures of the bypass mode was consistent with source sampling results conducted during the same period of time. Those results showed nearly a threefold increase in the NH_4^+.

A final set of solid analyses was performed to determine the NH_4^+ content of solids collected at various points in the heat exchanger. The results of these analyses and the sampling locations are presented in Table V.

The average NH_4^+ content of the 1st stage at an average temperature of 376°C is 41.2 ± 6.4 ppm. The second stage solids have an average temperature of 609°C but only 19.7 ± 3.6 ppm NH_4^+ in the solids. This represents a nominal 52 percent loss of NH_4^+ in the transition of the solids from 376 to 609°C. This temperature effect is also evident in the results of NH_4^+ analysis of solids collected at site G. The solids temperature here was 815°C and no NH_4^+ was detectable in the 0.1 N H_2SO_4 washes.

Gas samples, collected with the impinger sampling train with 0.1 N H_2SO_4 solutions were obtained at site E, F, and I. The results of these samples showed no free NH_3 at site E but from one sample acquired at site F, the second stage of the heat exchanger, a concentration of 6.2 $\mu g/m^3$ was detected. This indicates that solid phase NH_4^+ is being converted to gaseous NH_3 as the temperature of the solids increase. For the alkali bypass sampling site I, three gaseous NH_3 samples showed concentrations of 2.2, 0.8, and 1.1 $\mu g/m^3$ NH_4^+. The average concentration of the NH_4^+ at site I is 1.4 $\mu g/m^3$. This does not seem significant relative to an equivalent 4.4 and 16.0 $\mu g/m^3$ NH_4^+ measured at the baghouse emissions for the mill on and mill off modes respectively.

Because NH_4^+ was absent in the limestone and the NH_3 in the kiln gas emissions and because NH_3 was extracted from the raw shale, a study of the effect of temperature on NH_3 evolution from the shale was performed. As described in the Experimental Section, a preweighed sample of shale was placed in the heating device, and placed in the muffle furnace. Ammonia free air was pulled over the sample and through the impinger solutions as the sample was heated to the preset temperature. Initially, temperatures of a nominal 650°C were used and heating time varied to correspond to comparable temperatures at the stage 1–stage 2 points of the heat exchangers. The results of a series of such measurements

Table II. Presence of NH_4^+ in acidified Loesch Mill samples.

Sample date	Collection time	Sample wt (g)	μg NH_4^+ detected	ppm by weight
6/5	10AM	8.3	228	27.4
6/5	11AM	8.0	259	32.3
6/5	12PM	9.8	407	41.5
6/5	1PM	9.6	441	45.9
6/5	2PM	13.7	341	24.9
6/7	7AM	7.7	327	42.4
6/7	8AM	9.4	352	37.4
6/7	9AM	8.0	352	44.0
6/7	10AM	7.9	338	42.8
6/7	11AM	12.6	404	32.1
6/7	12PM	11.1	328	29.5
6/7	1PM	10.0	417	41.7
6/7	2PM	6.7	329	49.1
6/12	12PM	10.9	304	27.9
6/12	1PM	16.6	396	23.9

Table III. Presence of NH_4^+ in kiln feed solids.

Sample date (mode)	Collection time	Sample wt (g)	μg NH_4^+ detected	ppm by weight
6/5 (on)	11AM	11.1	449	40.4
6/5	1PM	12.4	382	30.8
6/7 (on)	7AM	8.9	324	36.4
6/7	9AM	9.7	404	41.6
6/7	11AM	7.4	399	53.9
6/7	12PM	6.2	338	54.5
6/12 (on)	1PM	10.8	468	43.3
6/6 (off)	7AM	8.9	324	36.4
6/6	9AM	9.7	404	41.6
6/6	11AM	7.4	399	53.9
6/6	1PM	6.2	338	54.5
6/11 (off)	11AM	18.0	513	27.1
6/11	1PM	18.9	552	29.2
6/13 (off)	7AM	15.1	563	37.3
6/13	9AM	18.6	636	34.2
6/13	1PM	15.7	636	40.5

Summary

A thorough study of the presence of and the likely source of NH_3 in a dry process Portland Cement manufacturing plant was conducted. Systematic analysis of the presence of NH_4^+ and gaseous NH_3 in the various locations of the process established a relationship between temperature and free NH_3 similar to Griffin, *et al.*[4]

Sampling fragments of the limestone shortly after a mine blast has shown that a minimum amount of NH_4^+ comes from the blasting powder. A more significant contributor was found to be the shale portion of the raw products.

Sampling for gaseous NH_3 in the emissions of the alkali bypass, an effective way of sampling the kiln gases, showed little NH_3 from the kiln. In contrast to Gartner *et al.*[5] that suggested nitrogen reduction and/or the production of NH_3 in the kiln clinker under reducing conditions, this cement kiln has little NH_3 associated with the combustion zone.

Heating the shale and collecting the liberated gases have shown that the shale is a major source of NH_3 and that it is likely liberated during the heating of the kiln feed. Analysis of the solids has shown that no NH_4^+ is present in the kiln feed after it has reached 815°C, a further indication that the NH_3 is liberated in the early stages of the heat exchanger.

Both the effects of heating the shale and collecting more NH_4^+ in the baghouse during the lower mill mode temperatures may offer a control strategy for controlling the detached plume.

Disregarding the engineering and/or economical aspects of such, preheating the shale before combining it with the limestone in the raw feed could greatly reduce the NH_3 in the plant's emissions. This NH_3 reduction in the emis-

are shown in Table VI.

The data in Table VI indicate that there is a lack of homogeneity in the shale with widely varying NH_3 evolution; the data also show that all of the NH_3 can be liberated from the shale at a minimum heating time of 1 hour at these temperatures. With the high value of 125.9 not included, an average of 51.4 ± 11.2 ppm of NH_4^+ can be evolved from the shale.

A second series of shale samples were heated in the same manner except at progressively lower temperatures. The result of the second series of runs is shown in Table VII.

As data in Table VII indicated, NH_3 evolution from the shale is nearly complete at a nominal 482°C for as little as an hour (94 percent). These data also indicate that the shale fraction of the kiln feed will emit NH_3 as it passes through the heat exchanger.

To confirm that NH_4^+ detected in the impinger was due to NH_3 liberation and not conversion of nitrate or nitrite, several samples with known amounts of the nitrogen oxides were heated. Volumes of solutions containing nitrate or nitrite salts were added to shale residue samples from which all NH_3 had been liberated due to heating. These residue samples with the known additions were then heated as the original shale samples and the 0.1 N H_2SO_4 impinger catches analyzed for NH_4^+.

Table V. Presence of NH_4^+ in heat exchanger solids.

Sampling site	Sample temperature (°C)	Sample wt (g)	μg NH_4^+ detected	ppm by weight
E	388	8.1	287	35.4
E	339	12.9	480	37.2
E	377	9.0	479	53.2
E	387	7.8	361	46.3
E	382	8.0	347	43.3
E	379	13.2	416	31.5
F	610	11.7	196	16.8
F	604	14.1	168	11.9
F	598	7.8	157	20.1
F	608	8.6	170	19.8
F	615	5.3	151	28.4
F	615	7.5	161	21.4
G	815	9.7	None	None
G	815	9.6	None	None

For three different samples containing 3000 μg of added nitrate each, a maximum of 163 μg of NH_4^+ was derived from heating the residues for 6 hours. For the three samples, an average of 57.5 μg of NH_4^+ was collected.

For three different samples containing 3000 μg of added nitrite each, a maximum of 102 μg of NH_4^+ was derived from heating the residues for 6 hours. For the three samples, an average of 52.7 μg of NH_4^+ was collected.

Table IV. Presence of NH_4^+ in baghouse solids.

Sample date (mode)	Sample temperature (°C)	Sample wt (g)	μg NH_4^+ detected	ppm by weight
6/5 (on)	172	5.7	763	134
6/5	172	5.0	725	145
6/5	172	4.9	778	159
6/5	172	6.4	663	104
6/12 (on)	168	6.5	667	103
6/6 (off)	252	8.5	600	71
6/6	252	5.7	524	92
6/6	252	6.2	451	73
6/6	252	6.3	493	78
6/11	249	6.0	313	52

Table VI. Evolution of NH_3 from shale.

Sample wt (g)	Temperature (°C)	Heating time (h:min)	NH_4^+ (μg)	μg NH_3 / g sample	Total
63.6	677	3:17	2997	47.1	
63.6	677	2:00	0	0	47.1
67.8	671	2:48	2956	43.6	
67.8	670	3:27	12	0.2	43.8
73.1	660	3:43	2860	39.1	
73.1	655	3:28	15	0.2	39.3
69.8	672	3:12	3662	52.4	
69.8	655	3:18	5	0.1	52.5
65.5	655	2:05	2362	36.1	
65.5	655	0:58	37	0.6	36.7
67.8	649	2:08	8532	125.8	
67.8	654	0:58	10	0.1	125.9
64.3	655	1:00	3242	50.4	
64.3	656	1:46	40	0.6	51.0
64.3	667	1:27	5782	89.4	
64.3	659	2:00	11	0.2	89.6

Table VII. Evolution of NH_3 from shale.

Sample wt (g)	Temperature (°C)	Heating time (h:min)	NH_4^+ (μg)	μg NH_3 / g sample	Total
64.7	588	2:55	5782	89.4	
64.7	583	2:53	11	0.2	89.6
64.4	604	1:00	4402	68.4	
64.4	588	2:53	117	1.8	70.2
62.4	593	2:11	6122	98.1	
62.4	593	2:50	32	0.5	98.6
67.8	491	2:52	2967	43.8	
67.8	478	3:20	41	0.6	44.4
63.7	510	1:15	2742	43.0	
63.7	493	2:53	168	2.6	45.6
64.1	488	1:59	2172	33.9	
64.1	485	2:36	84	1.3	35.2

sion would result in a reduction of NH_4Cl formed as the emissions cool thus reducing the opacity of the detached plume.

Equally possible would be to drop the temperature of the gas stream sufficiently to form NH_4Cl before the baghouse. This approach would enhance the formation and collection of particulate NH_4Cl prior to the stack exit. Returning the baghouse solids to the storage silos would be questionable as a cyclic buildup of NH_4Cl would eventually override the goals of the effort. A more practical solution at this plant would be to feed the baghouse solids collected at the lower temperature to an adjacent wet process where no NH_3 problems exist.

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